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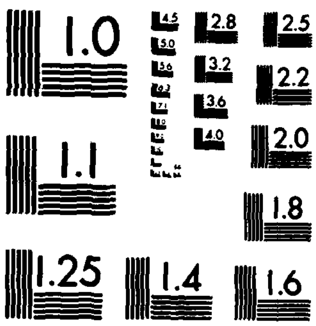

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GC-P-81-261

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DURING
ACCIDENT AND POST-ACCIDENT CONDITIONS

GEO-CENTERS, INC.

(12)

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BEHAVIOR OF CHARCOAL FILTERS
DURING
ACCIDENT AND POST-ACCIDENT CONDITIONS

UNSOLICITED PROPOSAL
PREPARED FOR
THE UNITED STATES
NAVAL RESEARCH LABORATORY
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CONTENTS

FIGURES

TABLES

INTRODUCTION

The widespread use of charcoal filters for air purification in nuclear environments dictates that a large knowledge base be accumulated on the performance of these materials. The radioactive isotopes of iodine are present as a by-product of U^{235} fission and these present a significant health hazard if not removed from the atmosphere. There are regulatory criteria enforced by the Nuclear Regulatory Commission directing the removal of airborne isotopes.

The residual life test of commercially available charcoal filters used in research and in commercial reactor operations, and those reactors used for military applications (such as modern vessel propulsion systems), has not been fully established. This lack of information is especially deficient in the area of charcoal performance under accident and post-accident conditions.

Weathering conditions and composition of impregnated charcoals as well as radiation exposure are factors affecting filtering effectiveness. The filtering capability of a charcoal during high radiation levels and the long term effects of that radiation with respect to removal of airborne radioactive contaminants have not been established.

The growing awareness of the problems concerning contamination by airborne radionuclides has established the need for the gathering of information involving filtering effectiveness of charcoals, both during and after high levels of radiation exposure. This includes filters currently in use in reactor environments and also in clean-

room applications. This proposal deals with the design of experiments and equipment necessary to observe the performance of charcoal filters during accident and post-accident conditions, with variation of charcoal composition, weathering, damage to filters, and radiation exposure at peak level of 10^9 rads for accident iodine build-up on adsorber.

STATEMENT OF WORK

Geo-Centers, Inc. proposes a twelve-month effort to evaluate the performance of new and weathered charcoal when subjected to radiations of selected intensities and doses. Specifically, Geo-Centers proposes the following tasks:

1. Observe the performance of charcoals under accident and post-accident conditions, and determine the Methyl-I penetration as a function of the charcoal composition, weathering status, and level of radiation.

2. Make static exposures of charcoals to various radiation levels known duration, and analyze each carbon for Methyl-I^{131} penetration.

3. Make dynamic measurements on charcoals during the exposure of the charcoals to various radiation levels using an air flow containing Methyl-I^{127} and subsequently determine the penetration of Methyl-I^{131}

Portions of this research will be performed at the Naval Research Laboratory on a non-interfering basis. Results will be summarized in a final report.

TECHNICAL DISCUSSION

There is a critical need to remove low level concentrations of radionuclides that originate within nuclear power and fuel processing facilities. Radioactive iodine, krypton, and xenon not only present biologically harmful effects on humans exposed to these materials, but also detrimental effects on equipment resulting from radioactive contamination as well.

Filtering systems have been designed to limit the release of these contaminants during normal operational periods, and also in case of possible accidents. These systems incorporate charcoal filters which adsorb the contaminants present. Krypton, xenon, and elemental iodine do not present serious problems in removal, but secondary products such as methyl iodide (CH_3I) are more difficult to remove. The retention of iodine and methyl iodide by charcoals may be improved by impregnation with an amine or potassium iodide. The charcoal, which is not necessarily inert, has an extremely large surface area available for gas flowing through the filter. Degradation of the filter over time may involve either the charcoal or the impregnant alone, but usually degradation occurs at the gas/solid interface. The charcoal may also have a significant part in the performance of the impregnated adsorbent.

Air pollutants have a detrimental effect on the long time retention efficiency of iodine by adsorbent charcoals. The emission on NO_x in the reactor containment space and the formation of ozone combine to irreversibly degrade the intricate systems of microporosity that constitute the major fraction of adsorbing

surface area. The performance of these filters subjected to high levels of radiation for extended periods has yet to be determined.

Prior research on the trapping of radioactive iodine by charcoal filters was designed to simulate pre-accident conditions.[(1)] Studies included such factors as the effects on behavior by air pollutants, relative humidity, dew point, and organic solvent contamination. Some of these factors showed a marked effect on the penetration of radioactive methyl iodide (CH_3I^{131}) through the impregnated charcoal.

The high concentration of I^{131} above background levels in an accident situation would increase the reaction rate of iodine with the charcoal and the impregnated material. The ionization of the carbon network with I^{131} would be enhanced by probable radiation insults to the carbon and impregnant. The formation of organic - I^{131} compounds in the carbon bed would result subsequently in the "bleeding" of I^{131} into the effluent air at a rate which would be strongly dependent upon the existing weather damage to the charcoal and charcoal impregnant. The charcoal filters should behave in a manner such that they may be characterized by the many facets of degradation during weathering and by the magnitude of the radiation damage in a simulated accident. A systematic examination of the factors concerning iodine penetration will contribute to the understanding of many accident scenarios.

The systematic investigation of charcoal performance would include both static and dynamic testing of filters. The static measurements would involve exposing the filters to various radiation levels of fixed duration. Following the static exposure of the filters, the penetration of Methyl-I^{131} would be determined. Dynamic testing would entail the use of a system which allowed an air flow containing Methyl-I^{127} to be passed through a charcoal exposed to various radiation levels for a specified amount of time, and determining the penetration of Methyl-I^{131} after this

exposure. The sources of radiation to be used in the investigation would be a 1 Mev Cobalt-60 source and a 45 Mev Linac accelerator, both of which exist at the Naval Research Laboratory.

The equipment necessary to complete the static and dynamic tests of charcoals subjected to radiation in either of these sources would include the design and construction of appropriate containers for the charcoals, a source of air which has the capability of modification of the relative humidity, a standardized source of Methyl-I^{127}, and a portable chromatograph using a calibrated electron capture detector. A sufficient amount of stainless steel tubing would also be needed to reach the carbon sample which is placed under 12 feet of water. Determination of Methyl-I^{131} after radiation insult would be determined by the proposed ASTM method.(2)

By using data that has already been established for air quality measurements(3,4) and similar measurements taken simultaneously during weathering experiments, the effects of unmodified air flows through charcoal beds can be established. The effects of singular contaminants such as NO_x, SO_x, CO, hydrocarbons, and water vapor or some combination of any of these components may be determined by using mixtures of bottled gases fed through a mixing chamber into the charcoal sample.

After the weathering procedure has been completed for a predetermined time, the irradiation of the charcoals may be started. Experiments based on non-radioactive Methyl-I^{127} and weathering conditions in non-accident modes(5,6) have led to the conclusion that there may be significant degradation of the carbon matrix. Figure 1(6) shows the effect of humidification on a new charcoal BC - 727 (coconut - charcoal impregnated with KI_x). Pre-humidification is typically several tens of hours with 95% RH and flow rates dependent on the size of the filter. It can be clearly seen from Figure 1 that the effect of prehumidification of a new charcoal can be a reduction of that filters' ability to remove Methyl-I^{127} from

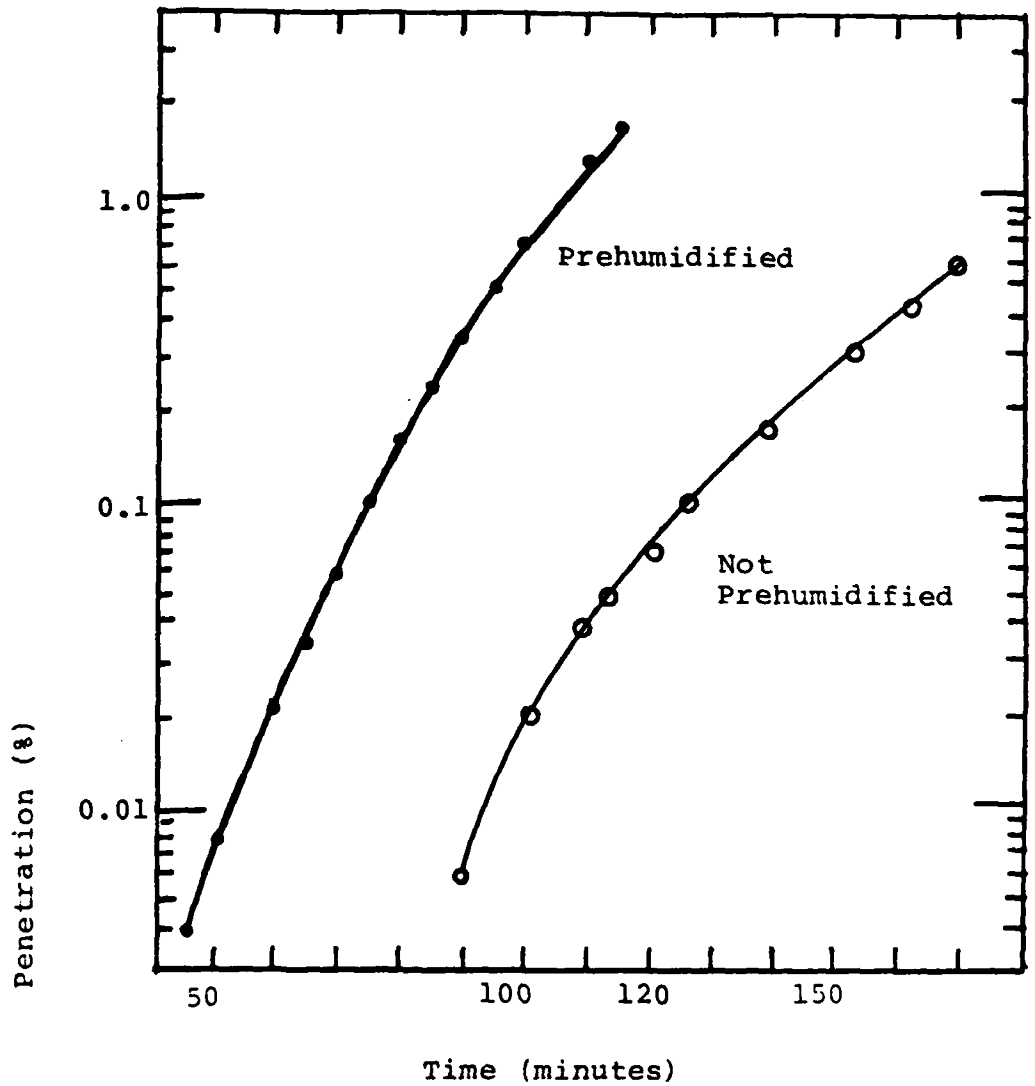

Figure 1. Effect of prehumidification on penetration of Methyl-I^{127} through charcoal (BC-727). Air flow at 90% RH.

the atmosphere when compared to a non-prehumidified charcoal exposed for the same time period. The use of outdoor air in a filter for three months reduces its ability to remove iodide by a factor of 5 after 120 minutes, as can be seen from Figure 2 (NACAR G-615 (KI + triethylenediamine impregnant)). In general, the effect of prehumidification is to reduce the trapping efficiency; the relative humidity of the air flow is also important in determining penetration. Air flows at low relative humidity increase the trapping efficiency of charcoal filters.

In an accident situation, the ability of a filter to retain radioactive iodide compounds is of utmost importance. Table 1[6] shows data taken for a typical charcoal which had seen some service from utility operations.

Table 1[6]

Emission of CH_3I^{127} during Air Purge

Test	Location of Maximum Cover (min)	(mg/M^3)	Total Dur. (min)	Purge Total (mg)	Total Dose (mg)	Recovery (%)
1	170	3.20	1200	3.54	3.70	94
2	230	2.60	1300	3.34	3.34	93
3	1700	0.50	7000	5.48	7.92	69

Tests 1 and 2 from Table 1 were at 95% RH and test 3 was at 30% RH. The emission of Methyl-I^{127} in test 3 began slowly, and only obtained a maximum of 0.50 mg/M^3. While there is a high amount of emission of material at an RH of 30%, the average concentration of the effluent material is low because of the large volume of air required to return to a baseline level. For this exhausted charcoal, the amount of material released for the 95% RH case is very high, and the filter behaves more like a gas chromatograph. The effects seen by prehumidification on

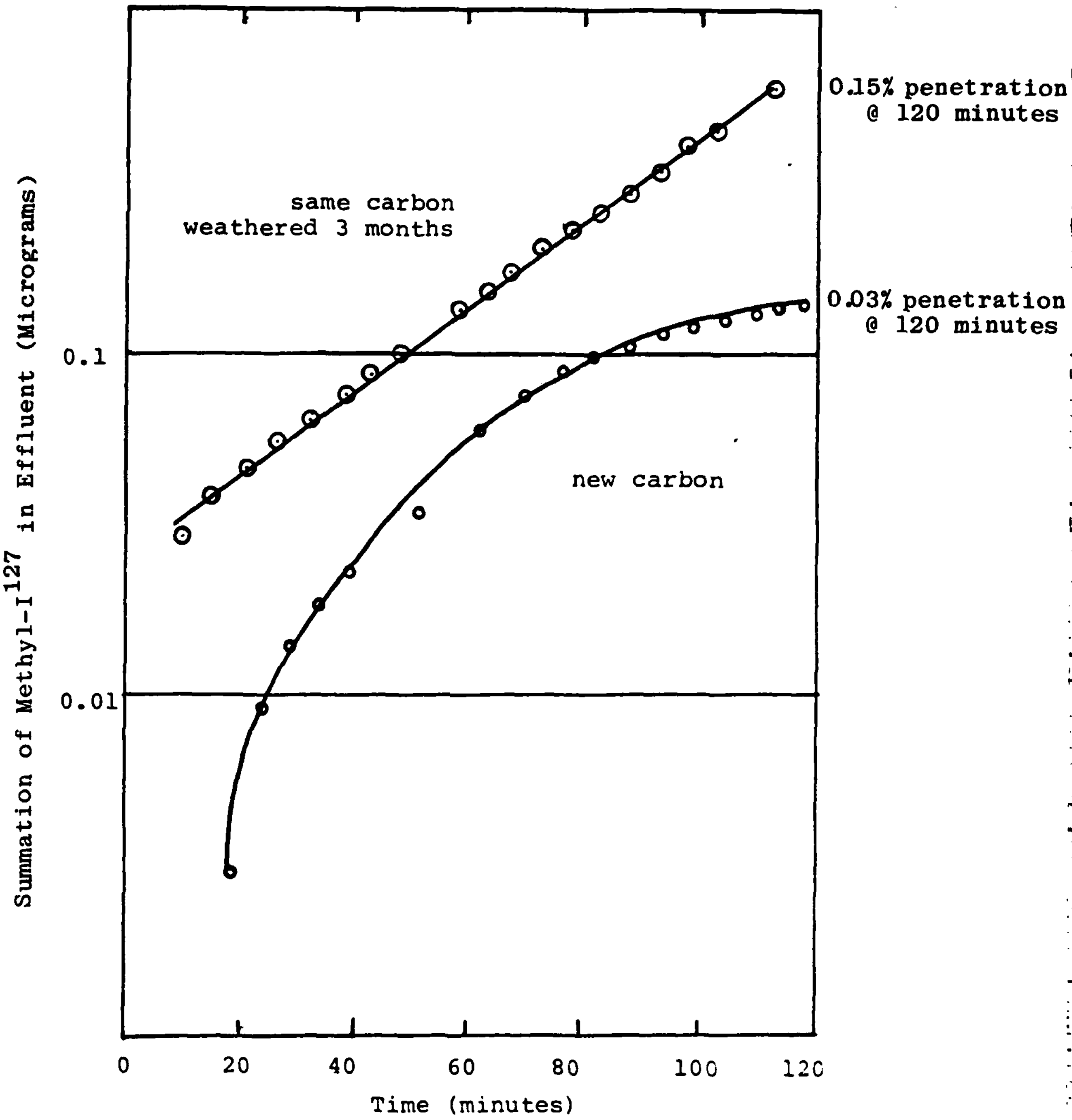

Figure 2. Effect of weathering on the in-line measurement Methyl-I^{127} on charcoal (NACAR G-615).

the behavior of new charcoals do not affect the amount of materials released from previously exhausted charcoal. High relative humidity and prehumidification appear to promote early iodide elution from the filter, which could prove to be significant considering the certainty of stream evolution during an accident scenario. Prolonged air flows do not appear to regenerate the charcoal since a second dose to the charcoal after purging show a much greater penetration than did the previous dose.

These effects will also be compounded by the ionization effects of intense radiation, but the magnitude of the damage induced by radiation will also be additive to the weathering effects, the greatest damage being inflicted by the amount of water vapor. Deterioration of the beds is also evident by exposure to ozone and sulfur dioxide. The summation of these effects under accident conditions are to be evaluated under this proposal.

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